Identifying the processes controlling the distribution of H_2O_2 in surface waters along a meridional transect in the eastern Atlantic

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[1] Hydrogen peroxide (H_2O_2) is an important oxidant for many bio-relevant trace metals and organic compounds and has potential as a tracer for mixing in near surface waters. In this study we combine H_2O_2 and bio-optical measurements with satellite data for a meridional transect from 46°N to 26°S in the eastern Atlantic in order to determine the key processes affecting its distribution. Surface H_2O_2 ranged from 21–123 nmol L^{-1}, with maximum inventories (0–200 m) of 5.5–5.9 mmol m^{-2} found at 30°N and 25°S. Analyses showed a strong positive correlation of surface H_2O_2 with daily irradiances and recent precipitation, though poor correlations with CDOM suggest sunlight is the limiting reactant for H_2O_2 formation. Vertical distributions of H_2O_2 were controlled by a combination of mixing processes and phytoplankton activity. The present study highlights processes controlling global H_2O_2 distributions and points towards the development of parameterization schemes for prediction via satellite data. **Citation:** Steigenberger, S., and P. L. Croot (2008), Identifying the processes controlling the distribution of H_2O_2 in surface waters along a meridional transect in the eastern Atlantic, *Geophys. Res. Lett.*, *35*, L03616, doi:10.1029/2007GL032555.

1. Introduction

[2] In marine systems H_2O_2 functions as a strong oxidant or a reductant and thus it is important for the cycling of organic compounds and trace metals like Fe [*Millero and Sotolongo*, 1989]. H_2O_2 is the most stable intermediate in the four-electron reduction of O_2 to H_2O and is mainly produced in the water column by photochemical reactions involving dissolved organic matter (DOM) and O_2 [*Yuan and Shiller*, 2001]. Light absorbed by DOM induces an electron transfer to O_2 forming O_2^-, which undergoes disproportionation to form H_2O_2. Typical open ocean H_2O_2 profiles show an exponential decrease from a surface maximum consistent with the downwelling irradiance. Maximum concentrations of 300 nmol L^{-1} have been reported in Equatorial and Tropical regions with high DOM concentrations as for the Amazon plume [*Croot et al.*, 2004]. In regions with low DOM and low sunlight, surface H_2O_2 levels are much lower with Southern Ocean values of 10–20 nmol L^{-1} [*Croot et al.*, 2005].

[3] Another potential source for H_2O_2 in surface seawater is from precipitation which preferentially removes H_2O_2 from the atmosphere during rain events [*Cohan et al.*, 1999], consequently the atmospheric input at the equator and in the Inter Tropical Convergence Zone (ITCZ) is high [*Croot et al.*, 2004; *Weller and Schrems*, 1993; *Yuan and Shiller*, 2000] compared to areas with less precipitation. H_2O_2 in the ocean is also produced biologically by phytoplankton [*Palenik and Morel*, 1988]. While photochemical production is considered the dominant pathway for H_2O_2 formation, in a few cases in the Southern Ocean, distinct H_2O_2 maximums at depth, corresponding to the chlorophyll maximum, suggest a significant biological source of H_2O_2 [*Croot et al.*, 2005].

[4] Removal pathways also determine H_2O_2 concentrations in the water column and include the Catalase and Peroxidase activity of phytoplankton [*Moffett and Zafiriou*, 1990] along with redox reactions with reduced metals (e.g. Fe(II) and Cu(II)) [*Millero and Sotolongo*, 1989; *Moffett and Zika*, 1987]. The 'dark decay life-time' of H_2O_2 can vary from hours to weeks in the ocean [*Petasne and Zika*, 1997], but typically is around 4 days in the open ocean [*Plane et al.*, 1987]. Overall, the decay rate of H_2O_2 is apparently controlled by several factors including H_2O_2 concentration, colloid concentration, bacteria/cyanobacteria numbers and temperature, which controls enzymatic decay [*Wong et al.*, 2003; *Yuan and Shiller*, 2001]. Due to its short lifetime H_2O_2 shows potential as a tracer for recent vertical mixing activity [*Johnson et al.*, 1989].

[5] In the present study we compare H_2O_2 profiles with physical and bio-optical measurements and available satellite data to determine the major processes controlling the distribution of H_2O_2 in the upper ocean along a meridional transect in the eastern Atlantic.

2. Methods

2.1. Sampling

[6] Samples were collected during the GEOTRACES cruise, ANTXXIII-I from 14 October to 17 November 2005 on board the German research vessel *R. V. Polarstern* on a transect between Bremerhaven and Cape Town. Six to seven depths were sampled for H_2O_2 from the upper 200 m at 19 stations (Figure 1), at local noon, using Niskin bottles on a standard CTD rosette. All analytical work was carried out in an AirClean class 100 laminar flow clean bench. Chlorophyll and chromophoric dissolved organic matter (CDOM) were measured in samples collected from the same CTD/rosette cast.

[7] By sampling only at local noon we were unable to examine the importance of the solar driven diel cycle in H_2O_2, by which variations of up to 40 nmol L^{-1} H_2O_2 have been reported with maxima in the afternoon or early evening [*Yuan and Shiller*, 2001; *Zika et al.*, 1985a; *Zika*

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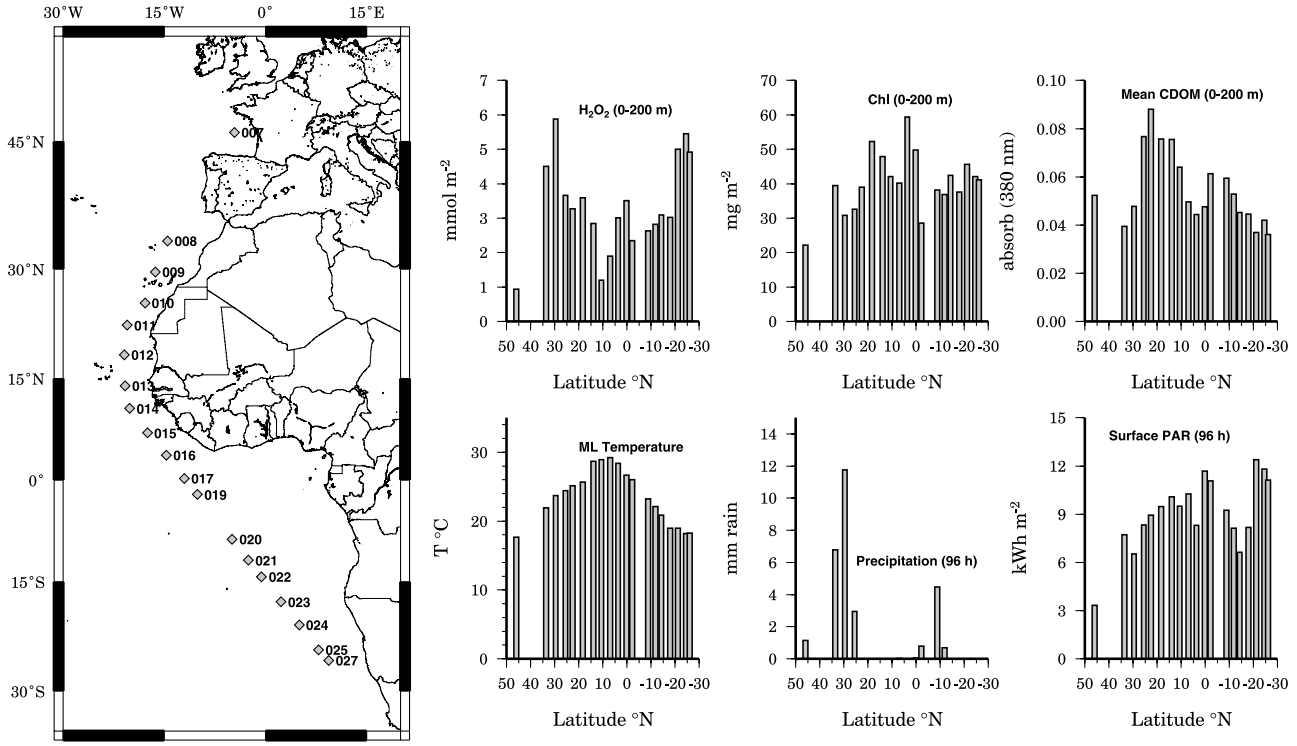

Figure 1. (left) ANTXXIII-1 cruise transect with location of the sampling stations. (right) Representative latitudinal distributions of some of the analysed parameters along the ANTXXIII-1 cruise transect. Clockwise from top left: H_2O_2 inventory (0–200 m), chlorophyll inventory (0–200 m), mean CDOM in the upper 200 m, surface PAR for the preceding 96 hours (source: HelioClim-2 database), precipitation for the preceding 96 hours (source: TRMM data), and mixed layer temperature.

et al., 1985b]. In the present work by sampling at the local noon, we are able to provide a valid comparison between stations along the transect but it is clear more work on the diel cycling of H_2O_2 is required.

2.2. H_2O_2 Measurements in Surface Waters

[8] Samples were drawn into 100 mL low density brown polyethylene bottles which were impervious to light. Unfiltered samples for H_2O_2 were analyzed within 1–2 h of collection using a flow injection chemiluminescence (FIA-CL) reagent injection method [*Yuan and Shiller*, 1999] as described by *Croot et al.* [2004]. Five replicates of each sample were analyzed with a typical precision of 2–3% in the concentration range of 2–120 nmol L^{-1} and a detection limit (3σ) of typically 0.6 nmol L^{-1}.

2.3. Measurement of the Natural Light Field Within the Upper Water Column

[9] A freefalling spectroradiometer (SPMR, Satlantic) was deployed for measuring the natural light field within the upper water column (down to 150–200 m). The spectral downwelling irradiance was measured at 13 wavelengths covering a spectral range from 339–682 nm.

2.4. Photosynthetically Active Radiation (PAR) Data

[10] Hourly sub-surface PAR (400–700 nm) estimates for the sampling period were obtained from the Helio-Clim-2 database (http://www.soda-is.com/eng/services/service_invoke/gui.php) which is constructed from METEOSAT data using the Heliosat-2 method [*Rigollier et al.*, 2004].

2.5. Measurement of Chlorophyll and CDOM Within the Upper Water Column

[11] The samples were filtered to collect the particulate matter and then stored in liquid nitrogen. Samples were analyzed post-cruise with HPLC (High Performance Liquid Chromatography) by R. A. Reynolds and D. Stramski (Scripps Institution of Oceanography, U.S.). Spectral absorption measurements of CDOM at 326 and 380 nm were made onboard the ship by R. Röttgers (GKSS Research Centre, Germany) using PSICAM [*Rottgers and Doerffer*, 2007].

2.6. Other Parameters

[12] Salinity, temperature and transmission were measured via a CTD (SBE 911plus, Sea-Bird Electronics). The integrated (over 3 h) precipitation data in mm were obtained from NASA TRMM (Tropical Rainfall Measuring Mission) product 3B42 using the GIOVANNI web-interface (http://daac.gsfc.nasa.gov/techlab/giovanni/).

2.7. Statistics

[13] A Spearman rank test was performed on the data which yielded pairwise correlation coefficients (ρ) between the parameters. All statistical analyses were done with SigmaStat 3.1 (Systat Software Inc.).

3. Results

3.1. Latitudinal Patterns of H_2O_2, Irradiance, SST, Chlorophyll/CDOM, and Precipitation During ANT XXIII-1

[14] A transect from 46°N to 26°S across the Atlantic covers a wide range of upper ocean environments [*Sarthou*

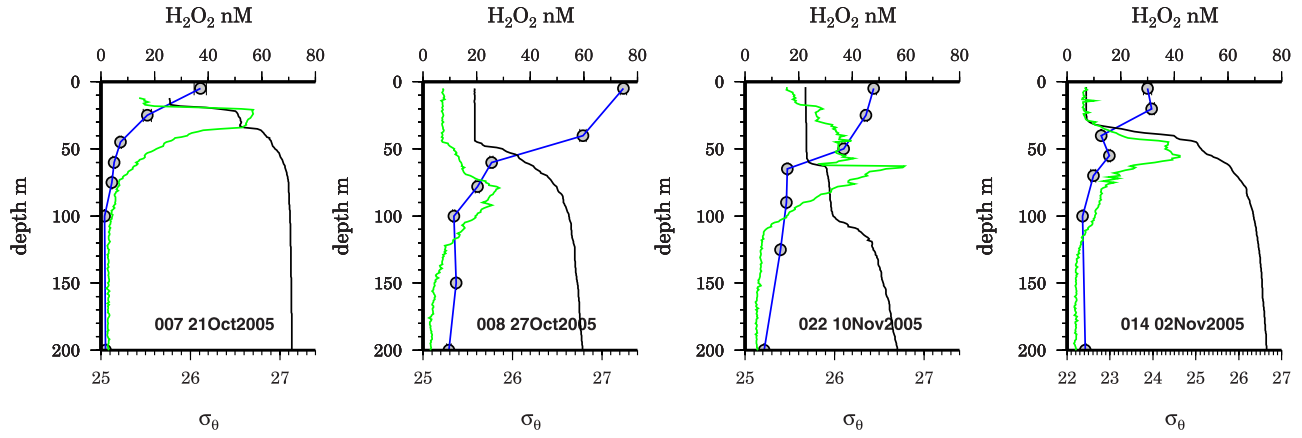

Figure 2. Vertical profiles of H_2O_2 (black circles on blue line), density σ_θ (black line), and chlorophyll fluorescence (green line, arbitrary fluorescence units where the x-axis spans from 0 to 1) showing (left) the characteristic exponential decrease (representing Stn PS 69/007, 10–13, 15–20, 24–27), (middle left) the effect of 7 mm precipitation (previous 96 h accumulated, PS 69/008), (middle right) the effect of vertical mixing (representing Stn 21–23), and (right) coinciding peaks in chlorophyll fluorescence and H_2O_2 (representing Stn 9, 14).

et al., 2003]. Surface chlorophyll concentrations ranged from 0.09–0.29 μg L^{-1}, the sea surface temperature (SST) ranged from 18–29°C with H_2O_2 inventories (0–100 m) ranging from 0.9–5.9 mmol m^{-2} (Figure 1). Highest H_2O_2 inventories between 5.5–5.9 mmol m^{-2} were found at stations 009 (30°N) and 025 (24°S), consistent with the earlier findings of *Yuan and Shiller* [2005], lowest inventories of 0.9–1.29 mmol m^{-2} at stations 007 (46°N) and 014 (10°N) (Figure 1). The highest integrated (0–200 m) chlorophyll values of 50–60 mg m^{-2} were found off Mauritania (Stn PS 69/012) and near the Equator (Stn 016) (Figure 1). CDOM absorbance, averaged over 0–200 m depth, was highest off north–west Africa (Stn 010–014) (Figure 1).

[15] Both the measured instantaneous PAR and the temperature of the mixed layer (Figure 1) reached maximum values near the Equator at station 016 and 15 respectively and decreased until station 021. Significant precipitation events (5–12 mm) occurring in the preceding 4 days before station occupation were detected for the stations off Morocco (Stn 008–009) and in the South Atlantic (Stn 021) (Figure 1).

3.2. Vertical Distribution of H_2O_2, Light, and Chlorophyll/CDOM During ANT XXIII-1

[16] The vertical profiles of H_2O_2 show the typical exponential decrease in the upper 50–100 m (Figure 2). At most of the stations H_2O_2 concentrations below 100 m depth were <10 nmol L^{-1}. Lowest surface concentrations (<30 nmol L^{-1}) were found in the Bay of Biscay (Stn PS 69/007) and at station 014 (10°N) coinciding with cloudy conditions. Maximum concentrations (123 nmol L^{-1}) were recorded in the southern Angola Basin (Stn 025) under clear skies. The vertical distribution of H_2O_2 in the Angola Basin (i.e. Stn PS 69/021-023) showed a deviation from the expected exponentially decrease, with almost constant H_2O_2 concentrations for the upper 50 m, followed by a strong decrease towards 60–80 m (Figure 2). At stations 009 and 014 small increases in H_2O_2 concentrations anomalous from the normal exponential pattern coincided with both the chlorophyll and CDOM maximums.

[17] Surface chlorophyll was elevated in the Bay of Biscay (Stn 007), off Mauritania (Stn 012), and in the Angola Basin (Stn 023). Vertical chlorophyll profiles showed subsurface maxima and sharp decreases with depth. Maximum surface absorbance of CDOM at 380 nm was observed in the Bay of Biscay (Stn 007) and off Mauritania (Stn 012). CDOM absorbance was low at the surface and reached highest values at the chlorophyll maximum and stayed high until 200 m depth. The average euphotic depth (1% PAR, z_{eu}) was 70 m (range 45–101 m) along the transect.

3.3. Statistical Analyses

[18] See Table 1. The vertical distribution of H_2O_2 was strongly correlated, as expected, with irradiance and was strongest at 442 nm ($\rho = 0.86$ $p < 0.05$ $df = 56$) and to a lesser extent with both PAR and UVA (340 nm). The vertical distribution of CDOM and H_2O_2 revealed a modest negative correlation ($\rho = -0.57$ $p < 0.05$ $df = 72$). H_2O_2 and chlorophyll depth distributions in turn were weakly correlated ($\rho = 0.32$ $p < 0.05$ $df = 75$). The depth of the chlorophyll maximum and the depth of maximum CDOM absorbance correlated strongly ($\rho = 0.76$ $p < 0.05$ $df = 15$, Stn 021–023 excluded due to strong mixing in the upper water column). A further strong correlation existed between vertical profiles of H_2O_2 and temperature ($\rho = 0.75$ $p < 0.05$ $df = 131$).

[19] A modest negative correlation was calculated for H_2O_2 inventories over the MLD and the average temperature over the MLD ($\rho = -0.56$ $p < 0.05$ $df = 18$), which becomes very strong ($\rho = -0.96$ $p < 0.05$ $df = 11$) excluding the stations of significant precipitation (Stn 7–10, 19–21). Strongly negative correlated were surface H_2O_2 concentrations and surface chlorophyll concentrations ($\rho = -0.75$ to -0.92). Modest and strong correlations ($\rho = 0.66$ and -0.74) were found between PAR and UVA irradiation respectively and surface H_2O_2 concentrations. Precipitation

Table 1. Results of Spearman Rank Analysis of Correlations Between H_2O_2 and Potentially Important Parameters[a]

	Irradiance						
	PAR	340 nm	442 nm	Water Temperature	Chlorophyll, HPLC	CDOM, absorbance at 380 nm	Precipitation $\geq$ 1mm
	H_2O_2 Surface Data						
ρ = ...	*0.28(0.66)*	*0.40(**0.74**)*	*0.29(0.48)*	−0.22 (−0.44)	**−0.75 (−0.92)**	*−0.28 (−0.11)*	**0.86**
p = ...	*0.25(<0.05)*	0.08(***<0.05***)	*0.22(0.11)*	0.37 (0.14)	**<0.05(<0.05)**	*0.24 (0.72)*	**<0.05**
df = ...	*18(11)*	18(***11***)	*18(11)*	18 (11)	**18 (11)**	*18 (11)*	**6**
	H_2O_2 Inventories						
	—	—	—	−0.56(**−0.96**)[b] < 0.05(**<0.05**) 18 (**11**)	*0.20 (0.01)*[c] *0.4 (0.96)* *18 (11)*	*−0.38 (−0.40)*[d] *0.11(0.19)* *18 (11)*	—
	H_2O_2 Depth profiles						
	0.71 (0.72) **<0.05 (<0.05)** **63 (40)**	**0.72(0.72)** **<0.05(<0.05)** **69 (42)**	**0.83(0.86)** **<0.05(<0.05)** **84(56)**	**0.75** (0.65) **<0.05** (<0.05) **131** (82)	0.32 (0.25) <0.05 (0.09) 75 (47)	−0.57 (−0.52) <0.05 (<0.05) 72 (47)	—

[a]Correlations measured are surface concentrations, water column inventories, and discrete concentrations at depth. Values in brackets were determined excluding stations of significant precipitation (Stn 7–10, 19–21).
[b]Temperature averaged over MLD, H_2O_2 integrated over MLD.
[c]Integrated over 0–200 m.
[d]CDOM absorbance averaged over 0–200 m, H_2O_2 integrated over 0–200 m.

and surface H_2O_2 values correlated strongly (ρ = 0.86 p < 0.05 df = 6) at the seven stations affected by rain. Excluding stations of significant rainfall generally increased the strength of the correlations in the surface data.

[20] Estimated noon sub-surface PAR values from the HelioClim-2 database were in good agreement with the measured instantaneous PAR (ρ = 0.70 p < 0.05 df = 18). Excluding the stations of significant precipitation (Stn 7–10 and 19–21) the sub-surface PAR estimates integrated over 24, 48 and 96 h before sampling correlated modestly with surface H_2O_2 (ρ = 0.66–0.84 p < 0.05 df = 11).

4. Discussion

[21] Our results show that the distribution and inventories of H_2O_2 observed during ANT XXIII-1 were affected by several factors related to variations in the sources, sinks and physical mixing at each location. H_2O_2 inventories were strongly negatively correlated to the SST in line with expected temperature effects on enzymatic sinks for H_2O_2 [*Wong et al.*, 2003]. Inventories of H_2O_2 decreased equatorwards in contrast to SST and daily PAR irradiance (Figure 1). The highest observed H_2O_2 inventories (0–100 m) were found at the start and near the end of the cruise (Figure 1) possibly reflecting a balance between photo-formation and temperature-dependent loss processes. The strong negative relationship between H_2O_2 and chlorophyll at the surface is probably related to increased Catalase and Peroxidase activity arising from increased phytoplankton biomass [*Moffett and Zafiriou*, 1990]. Latitudinal decreases in H_2O_2 suggest that sink terms are more sensitive to temperature than the photo-formation of H_2O_2 is to irradiance, a finding consistent with recent studies in the North–West Pacific [*Yuan and Shiller*, 2005]. Inventories of H_2O_2 decreased by 49% from station PS 69/009 to 016 despite an increase in irradiance of 49% (Figure 1), this may arise from the simultaneous increase in SST by 25% (Figure 1). However, there is a strong correlation of surface H_2O_2 and UV irradiance (340 nm) underlining the importance of UV radiation for the photo-formation of H_2O_2 [*Gerringa et al.*, 2004]. The modest to strong positive correlation of surface H_2O_2 with the preceding 24, 48 and 96 h total sub-surface PAR flux indicates that, away from areas of precipitation, there is a clear connection to the "light history" at each station. Since instantaneous irradiation data can not solely explain distribution patterns of H_2O_2 in seawater, databases such as HelioClim-2 are a useful new tool for estimating the "light history" in the Ocean.

[22] The vertical distribution of H_2O_2 was dominated by irradiance (Table 1) with most stations showing a characteristic exponential decrease (Figure 2). The correspondence of the chlorophyll and CDOM maximum with the local increases in H_2O_2 009 and 014 (Figure 2) suggest either; production of H_2O_2 by phytoplankton [*Croot et al.*, 2005; *Palenik and Morel*, 1988] or from fresh photo-labile phytoplankton exudates. Station 014 (Figure 2) shows the inverse shape of the depth profiles of CDOM and H_2O_2 causing the modest negative correlation between these parameters (Table 1) which is related to photobleaching of CDOM in surface waters [*O'Sullivan et al.*, 2005].

[23] At Station 008, high surface H_2O_2 levels show the effect of precipitation (Figures 1 and 2) on H_2O_2 levels. Although the number of rain events was small they correlated strongly with H_2O_2 surface concentrations (Table 1) confirming that rain is an important source for H_2O_2 to the surface ocean [*Croot et al.*, 2004]. In the Angola Basin (Stn 021–23), strong mixing in the upper water column with the euphotic depth (z_{eu}) and the MLD roughly equivalent, resulted in significantly higher H_2O_2 concentrations at depth than at other locations (Figure 2), highlighting the importance of upper ocean mixing processes [*Johnson et al.*, 1989] on H_2O_2 distribution. It should be noted however that strong mixing has no apparent effect on H_2O_2 inventories. The strong correlation between vertical profiles of temperature and H_2O_2 (Table 1) is not unexpected as both parameters are forced by sunlight and have similar vertical distributions, in our study region, with highest values at the surface and decreasing values with depth. Increased temperatures should shorten the lifetime of H_2O_2 [*Moffett and Zika*, 1987; *Wong et al.*, 2003] which is seen in the negative correlation between H_2O_2 inventories and SST.

5. Conclusion

[24] Measurements of H_2O_2 during ANT XXIII-1 show a number of factors influenced its distribution. The latitudinal

patterns observed indicate irradiance, water temperature and recent precipitation as key controls. Vertical distributions of H_2O_2 were strongly controlled by photo-formation and mixing processes in the upper water column. The recent irradiation history and phytoplankton activity appear to be the key sources and sinks in determining the observed H_2O_2 levels, with CDOM playing a minor role suggesting sunlight is the key limiting reactant in the formation of H_2O_2 in the Tropical and Sub-Tropical surface ocean. This work points to the future possibilities of developing satellite based estimations of H_2O_2 in the global ocean.

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